

Scaling behavior of temperature-dependent thermopower in CeAu_2Si_2 under pressure

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We report a combined study of in-plane resistivity and thermopower of the pressure-induced heavy fermion superconductor CeAu_2Si_2 up to 27.8 GPa. It is found that thermopower follows a scaling behavior in T/T^* almost up to the magnetic critical pressure $p_c \sim 22$ GPa. By comparing with resistivity results, we show that the magnitude and characteristic temperature dependence of thermopower in this pressure range are governed by the Kondo coupling and crystal-field splitting, respectively. Below p_c , the superconducting transition is preceded by a large negative thermopower minimum, suggesting a close relationship between the two phenomena. Furthermore, thermopower of a variety of Ce-based Kondo-lattices with different crystal structures follows the same scaling relation up to $T/T^* \sim 2$.

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I. INTRODUCTION

Thermopower (S) of Ce-based Kondo lattice systems (KLS) exhibits a variety of unusual features depending on the coupling strength (J) between Ce-4*f* and conduction electrons, which is of both fundamental and application interests [1, 2]. Particularly, as J is enhanced by external pressure (p), typical features in $S(T)$ from Kondo to intermediate-valence (IV) regime can be observed even in a single compound, examples of which include CeCu_2Si_2 [3], CeAl_3 [4], CeCu_2Ge_2 [5], CePd_2Si_2 [5], CeCu_2 [6], CeRu_2Ge_2 [7]. At $p = 0$, $S(T)$ of these materials displays a positive maximum at high T but a negative minimum at low T . The increase of p tends to upshift the whole $S(T)$ curve. Once J becomes large enough for Ce to be in an IV state, one broad positive $S(T)$ maximum of the order of k_B/e ($= 87 \mu\text{V/K}$) persists, often with a shoulder at low T [8]. Theoretically, while at high and intermediate T the S behavior is understood as resulting from the interplay between Kondo and crystal field (CF) effects [9–11], the low- T negative S remains mysterious.

However, since in the aforementioned cases the long-range magnetic order (if any) disappears at $p < 8$ GPa [5–7], the way in which $S(T)$ evolves with p remains largely unexplored for weak Kondo coupling (small J) regime. In this context, it is worth noting that we recently found a giant overlap in p between the magnetic and superconducting phases of CeAu_2Si_2 , which contrasts radically with the observations made on its sister compounds CeCu_2X_2 ($X = \text{Si, Ge}$) [12]. Strikingly, the magnetic order of CeAu_2Si_2 persists up to $p_c \sim 22$ GPa [12], which, together with the small magnitude of $|S|$ ($\sim 2 \mu\text{V/K}$) [13–15], places the compound far from magnetic instabilities. Thus, the high- p $S(T)$ measurement of CeAu_2Si_2 allows not only to look for possible differences with the normal-state properties of CeCu_2X_2 , but also offers a rare opportunity to study the p -evolution of the characteristic features in $S(T)$ starting from a small J .

In this paper, we present thermopower $S(T)$ and electrical resistivity $\rho(T)$ measurements on the very same CeAu_2Si_2 crystal at p up to 27.8 GPa. Thanks to its large p_c , a remarkable scaling behavior is uncovered: the $S(T)$ data for $p \leq 20.5$ GPa, when normalized by a proper factor, collapse onto a single curve when plotted against T/T^* , where T^* is the temperature at which the first sign change of S occurs with decreasing T . The comparison with the results of ρ measurements shows that the normalization factor and T^* follow nearly the same p -dependence as the $-\ln T$ slope of $\rho(T)$ and overall CF splitting Δ_{CF} , respectively. In the p -range where superconductivity (SC) is enhanced with p , a large negative minimum in $S(T)$ develops below T^* . Further on, it is shown that the scaling relation is commonly applicable to Ce-based KLS provided that the coupling strength J is small enough. These results are discussed in regard to existing theories and their possible implication for Ce-based heavy fermion SC.

II. EXPERIMENTAL

We grow CeAu_2Si_2 crystals from Sn flux as described elsewhere [12, 16]. A sample cut from a crystal with low residual resistivity $\rho_0 \approx 2 \mu\Omega\text{cm}$ is used for in-plane $S(T)$ and $\rho(T)$ measurements, which are carried out in a Bridgman-type anvil pressure cell in the temperature range $1.3 < T < 300$ K with lead (Pb) as the p -gauge [17]. A photograph of the high- p chamber is shown in S1 of the Supplemental Material [18]. Compared with the previous study [12], we succeed in further miniaturizing the setup by reducing the sample size, the p -cell thickness (d) and the cross section of the thermocouple (TC) wires. Due to the rapid relaxation of the thermal gradient $\Delta T \propto \exp(-1/d)$, where $d \approx 40 \mu\text{m}$, special care must be taken to position the TC wires as close as possible to the heater. Following Ref. [7], $S(T)$ of the TC wires are

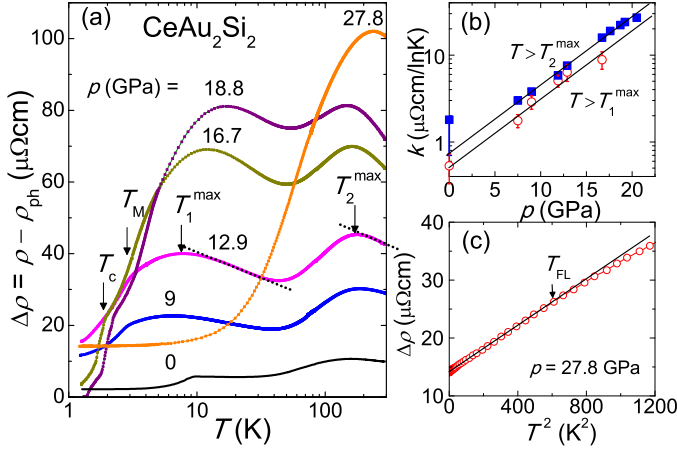


FIG. 1: (Color online) (a) Logarithmic T -dependence of the non-phononic contribution ($\Delta\rho$) to in-plane resistivity of CeAu_2Si_2 for typical pressures. The arrows indicate the two resistivity maxima at T_1^{\max} and T_2^{\max} , respectively, for 12.9 GPa. The two dashed lines evidence the $-\ln T$ dependence of resistivity. At 16.7 GPa, the magnetic and onset superconducting transitions are denoted by T_M and T_c , respectively. (b) The $-\ln T$ slopes k_1 and k_2 above T_1^{\max} and T_2^{\max} as a function of p . At $p = 0$, the large error in k_2 is due to the uncertainty in ρ_{ph} ; for the other pressures, the error is within the size of the symbol. The solid lines are a guide to the eyes. (c) $\Delta\rho$ at 27.8 GPa plotted as a function of T^2 . T_{FL} denotes the Fermi liquid temperature below which $\Delta\rho \propto T^2$ (solid line).

assumed to be p -independent, and we check that our results are free from their (possible) small variations under p . Corrections due to the misplacement of the TC wires are introduced by examining the chamber after depressurization. Within the experimental error of $\sim 20\%$, the results presented here are in good agreement with those obtained in another cell for the overlapping T -range ($1.3 < T < 7$ K) [19].

III. RESULTS AND DISCUSSION

Let us first briefly summarize the $\rho(T)$ results, which are essentially the same as in previous studies [12, 21]. Figure 1(a) shows the T -dependence of the non-phononic resistivity $\Delta\rho = \rho - \rho_{ph}$ for typical pressures, where ρ_{ph} is a phonon term derived from $\rho(T)$ of LaPd_2Si_2 [20] and assumed to be p -independent. Note that such a ρ_{ph} is a better approximation at low T than the linear one used previously [12, 21], which overestimates the actual ρ_{ph} contribution. At an intermediate p , say 12.9 GPa, and as observed in numerous KLS, $\Delta\rho(T)$ exhibits two maxima at T_1^{\max} and T_2^{\max} . Moreover, above each maximum $\Delta\rho(T) \propto -\ln T$, reflecting the incoherent Kondo scattering of the ground state and excited CF levels [22]. The increase of p has little effect on T_2^{\max} , but enhances both T_1^{\max} and the $-\ln T$ slope $k_i = -d(\Delta\rho)/d(\ln T)$ for $T > T_i^{\max}$ ($i = 1, 2$) [23]. Strikingly, as shown in Fig.1(b), k_1

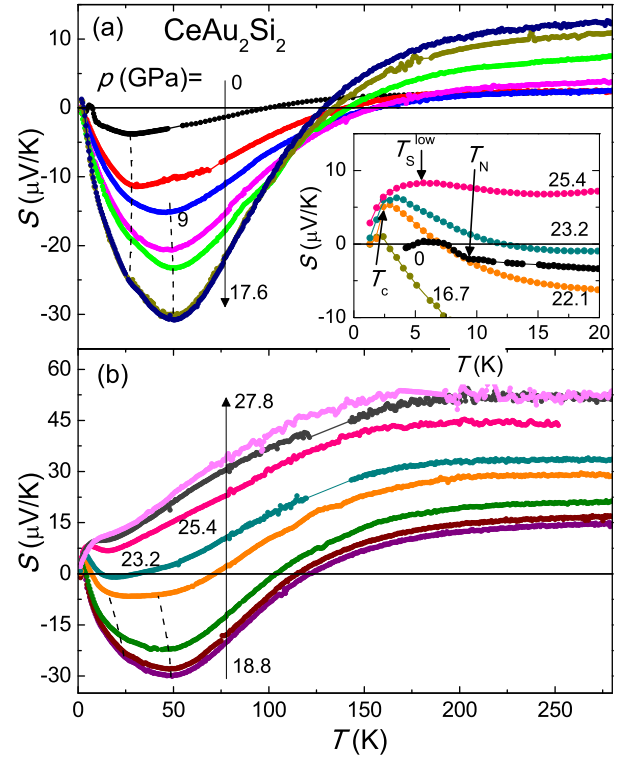


FIG. 2: (Color online) In-plane $S(T)$ of CeAu_2Si_2 for (a) $p \leq 17.6$ GPa and (b) $18.8 \leq p \leq 27.8$ GPa. The vertical arrows and dashed lines are a guide to the eyes. The inset shows the low temperature data for typical pressures. The temperatures corresponding to the superconducting transition and the maximum in thermopower are marked by arrows.

and k_2 share almost the same p -exponential dependence up to 16.7 and 20.5 GPa [24], respectively, pointing to a p -independent ratio of $k_2/k_1 \approx 1.5$. It must be emphasized that the p -dependence of the k_1 and k_2 slopes is practically independent of the choice of ρ_{ph} , but their ratio (k_2/k_1) can vary from 1.5 to 2.5 for different ρ_{ph} terms. Well below T_1^{\max} , signature of p -induced SC is detected at 16.7 and 18.8 GPa in addition to magnetic ordering, though the transition is broadened likely due to p -gradient. At the highest p of 27.8 GPa, Fermi liquid (FL) behavior extends up to $T_{FL} \approx 25$ K [Fig. 1(c)], and the Kondo temperature $T_K \approx 240$ K, estimated from the T_1^{\max} [25], is ~ 140 times the ambient- p value (1.7 K) deduced from the neutron scattering experiments [26].

We will now discuss the in-plane $S(T)$ data. At $p = 0$, $S(T)$ of CeAu_2Si_2 undergoes a sign change at $T^* \approx 100$ K and shows a minimum at $T_{\min} \approx 25$ K. This nonmonotonic T -dependence is in stark contrast to that found in polycrystalline non-magnetic LaAu_2Si_2 [15], suggesting that the dominant contribution to $S(T)$ already stems from weak Kondo scattering. As can be seen in Fig. 2(a) and (b), the evolution of $S(T)$ at low T is qualitatively different at p below and above 17.6 GPa. With increasing p below 17.6 GPa, it is observed for the first time

that the magnitude of the negative $S(T)$ is boosted from small to giant values typical of KLS. Concomitantly, T_{\min} increases to ~ 50 K for $p \geq 9$ GPa. On closer examination, a bump at ~ 25 K is still discernible, suggesting that there are actually two superimposed negative contributions (see dashed lines in Fig. 2). At 17.6 GPa, the S_{\min} value $\sim -30 \mu\text{V/K}$ is close to that of CeCu_2Si_2 at $p = 0$ [3, 25], an expected result given that at this p the volume of CeAu_2Si_2 is reduced to that of CeCu_2Si_2 [12]. Since the overall $S(T)$ behavior of CeCu_2Si_2 is very similar along the ab plane and c -axis with nearly isotropic S_{\min} [27], a weak anisotropy in $S(T)$ is expected for CeAu_2Si_2 around this p . In addition, it is worth mentioning that, after a partial depressurization from 27.8 down to ~ 17.6 GPa, while T^* is almost unaffected, the $S(T)$ magnitude is reduced concomitantly with an increase of ρ_0 , in line with a Nordheim-Gorter type relation [28]. Let us note the weak p -dependence of the temperature T^* , to which we will return below.

For $p \geq 18.8$ GPa, the trend in $S(T)$ versus p is reminiscent of previous observations in Ce-based KLS [3–7]. While the positive $S(T)$ keeps growing, the negative contribution decays and finally vanishes for $p > 23.2$ GPa. In fact, $S(T)$ starts to change back to positive value at low T already for $p \geq 16.7$ GPa. Following the sign change, $S(T)$ increases with decreasing T and then drops to zero due to the superconducting transition. However, since such sign change may already occur below 1.3 K at lower p , its detailed study is beyond the scope of the present paper. Incidentally, the anomaly in $S(T)$ associated with magnetic ordering is only observed at $p = 0$.

At room T , S at 27.8 GPa ($\sim 50 \mu\text{V/K}$) is ~ 20 higher than at $p = 0$. By contrast, there is only a fivefold enhancement in ρ . Together, these results signify a giant p -induced increase (~ 80 times) in the power factor S^2/ρ .

It is known that $S(T)$ of dilute Kondo alloys with $3d$ -impurities, when normalized by a factor of $1/S_N$, follows a universal function $f(T/\Theta)$, where the temperature Θ characterizes the coupling between the impurity local moments and the conduction electrons [29]. A similar situation is expected concerning $4f$ impurities, although no experimental observations have been reported to date [1]. The above $\rho(T)$ results which clearly demonstrate that, CeAu_2Si_2 behaves as a Kondo alloy above the temperature T_i^{\max} over a broad p range, leads us to examine whether such a scaling exists in this compound.

As shown in the main panel of Fig. 3(a), it turns out that for $T \geq T^*$ ($S \geq 0$) and p up to 20.5 GPa, the normalized S/S_N data fall on a single curve when plotted as a function of $t = T/T^*$. Note that, in our case, S_N is set as $S(1.79T^*)$, which is the same as, or very close to, the value of $S_{280\text{K}}$ for different p . At $t > 1.8$, the scaling curve can be fitted by the empirical formula $S \sim T/(T + T^*)$ for dilute Kondo systems [30], indicating that $S(T)$ is ascribed to the incoherent Kondo effect at sufficiently

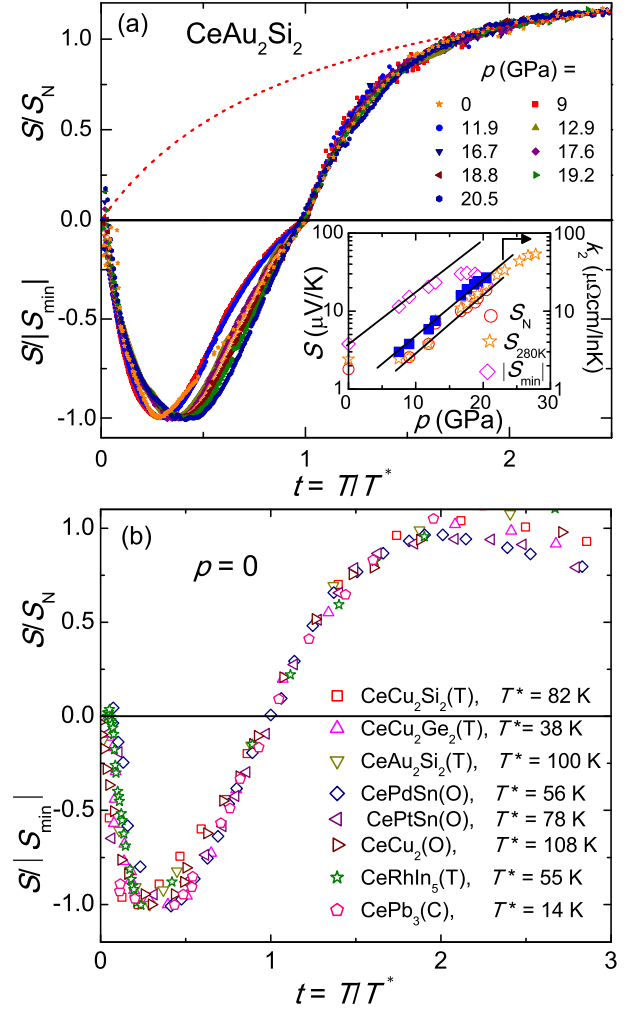


FIG. 3: (Color online) (a) The normalized $S(T)$ data for $p \leq 22.1$ GPa as a function of T/T^* . Data at 7.5 GPa are not considered due to possible gasket relaxation after initial pressurization. The red dashed line denotes the fit from an empirical formula for dilute Kondo systems (see text). The inset shows the p -dependence of the S_N , $S_{280\text{K}}$, and $|S_{\min}|$ and k_2 slope above T_2^{\max} . Note that both vertical axes are in logarithmic scale, and the solid lines are a guide to the eyes. (b) the normalized $S(T)$ versus T/T^* for a number of Ce-based Kondo-lattice compounds at ambient pressure. The letter in parentheses denotes the crystal symmetry, with T for tetragonal, O for orthorhombic, C for cubic.

high T . At $t < 1$, where $S(T)$ is likely governed by the thermal depopulation of the two upper CF doublets, a scaling is found when plotting $S/|S_{\min}|$ instead of S/S_N against t . The quality of the data collapse is less good than for $t > 1$, probably due to the interference of the two contributions to the negative $S(T)$ minimum mentioned above. For $p \geq 22.1$ GPa, no such scalings are observed at either low or high T , which we ascribe to the delocalization of Ce- $4f$ electrons [12].

The inset of Fig. 3(a) shows the resulting S_N , $S_{280\text{K}}$, and $|S_{\min}|$ plotted as a function of p , together with the

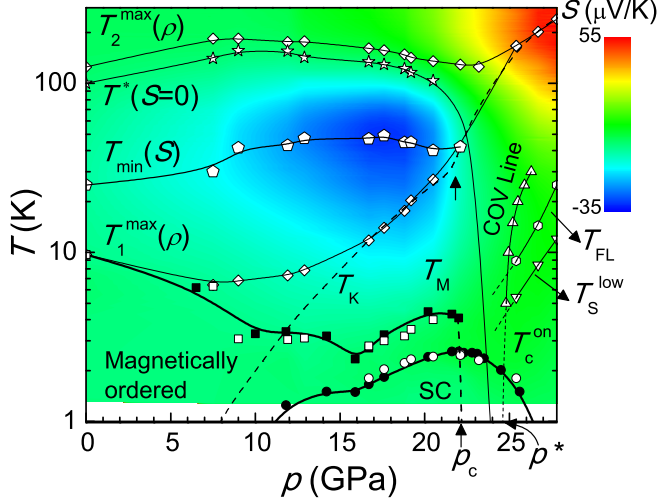


FIG. 4: (Color online) p - T phase diagram of CeAu_2Si_2 determined from the combination of resistivity and thermopower data. A contour map of the thermopower data is also included. Closed symbols represent the results from Ref. [12]. T_K is calculated from $T_K \sim \exp[-\frac{1}{NJn(E_F)}]$ (see text). The determination of T_{FL} and the crossover (COV) line can be found in the Supplemental Material [18]. Note that the vertical axis is in logarithmic scale.

slope k_2 for comparison. It is striking that S_N , S_{280K} and k_2 exhibit the same exponential dependence on p for $9 \leq p \leq 20.5$ GPa, and so does $|S_{\min}|$ for $p \leq 9$ GPa. According to Ref. [22], $k_2 \propto n^2(E_F)|J|^3$, where $n(E_F)$ is the density of the states at Fermi level. Assuming a p -independent $n(E_F)$, we have $S_N \sim S_{280K} \propto |S_{\min}| \propto |J|^3$ over a given p -range, which provides strong evidence that, at both low and high T , the $S(T)$ magnitude enhancement is due to the increase of J . In the high- T limit, this is consistent with the theoretical calculation of Kondo $S(T)$ for non-interacting f -electron spins [31]. Above 9 GPa, $|S_{\min}|$ deviates from the exponential behavior and tends to saturate, reflecting the competition between the positive and negative $S(T)$. Similarly, S_{280K} saturates above 26.7 GPa on approaching the value of k_B/e .

In Fig. 3(b), we compare the scaled $S(T)$ at $p = 0$ of various Ce-based KLS, including CeAu_2Si_2 , CeCu_2 [6], CeCu_2Si_2 [27], CeRnIn_5 [27], CeCu_2Ge_2 [32], CePb_3 [32], CePdSn [33], and CePtSn [33]. One can see that all data sets collapse on the same curve up to $t \sim 2$, despite the fact that T^* changes by nearly one order of magnitude. This trend exists for CeAl_2 [34], CeAl_3 [35], CeCu_5Au [36] and even β -Ce [37], and this list being by no means exhaustive. These results clearly demonstrate that the scaling of $S(T)$ is widely applicable to Ce-based KLS when J is sufficiently small, independently of the crystal structure or the local environment of the Ce ions. Furthermore, as found in CeAu_2Si_2 , it is expected that such a scaling still holds for these systems within a certain p -range. For example, this is the case of CeCu_2Ge_2 [5].

To gain more insight into the $S(T)$ evolution, we constructed a comprehensive p - T phase diagram (PD) of CeAu_2Si_2 by combining both $S(T)$ and $\rho(T)$ data, as shown in Fig. 4. We will first discuss the driving parameter T_K . As stated previously [12, 22], the temperature T_2^{\max} scales approximately the overall CF splitting Δ_{CF} , while T_1^{\max} gives an indication of T_K above 16 GPa when it rapidly grows to become much larger than T_M . With increasing p , the Kondo contribution to $\rho(T)$ at T_1^{\max} finally dominates over that at T_2^{\max} , and the system enters the IV regime with a trend to recover the full degeneracy of the Ce- $4f^1$ multiplet. At lower T , this trend is marked by the crossover (COV) line defined from the $\rho(p)$ drop due to the $4f$ electron delocalization [12, 18, 38]. On the other hand, $T_K(p)$ can be estimated as $T_K(p) \sim D \exp[-\frac{1}{NJ(p)n(E_F)}]$ [39], where D is the bandwidth, N is the degeneracy of the $4f$ -state and $J(p) = J_0 \times \sqrt[3]{10^{0.073p}}$ is obtained from the fitting of k_2 data in Fig. 1 (b). In order to qualitatively take into account the COV, N is assumed to increase linearly with p from $N = 2$ at p_c to $N = 6$ at 30 GPa. With the adjusted parameters $D = 700$ K and $J_0 n(E_F) = 0.05$, the calculated T_K reproduces reasonably well the T_1^{\max} for $p > 16$ GPa [40], confirming that T_K is increased by a factor of ~ 40 over the huge p -range of SC in CeAu_2Si_2 . Surprisingly, whereas no anomaly associated with the resistivity maximum at $T_1^{\max}(T_K)$ can be observed in $S(T)$ for $p < p_c$, this anomaly seems to be present in CeRu_2Ge_2 [7]. In fact, the above results strongly suggest that the Kondo effect is mostly reflected in the magnitude, rather than the T -dependence, of $S(T)$ in CeAu_2Si_2 below p_c .

For $p \geq 25.4$ GPa, the position of the high- T maximum in $S(T)$ agrees roughly with the T_1^{\max} of the resistivity maximum, and gives an independent estimation of T_K . By contrast, the temperature T_S^{low} of the low- T maximum is only half of T_{FL} and $\sim 5\%$ of T_K , where the T -dependent $\rho(T)$ term is still very small. Hence its origin is unlikely linked to the Kondo effect, but may be ascribed to short range magnetic correlations or to the crossover between the zero- T term S_0 [41, 42] and the usual Kondo term S_{mag} . To better clarify this issue, the sister compounds CeCu_2X_2 should be re-investigated, given that such maximum appears at a much lower p and hence can be studied in a more extended p -range [3, 5].

As already noted above, both T^* and T_{\min} vary weakly with p and behave very similarly to T_2^{\max} , unlike T_K , indicating that these quantities are controlled by Δ_{CF} . In this respect, the two terms contributing to the negative $S(T)$ minimum might correspond to the two excited CF levels at ~ 190 and ~ 250 K [43], each of them producing a minimum at $T_{\min} \sim \Delta_{CF}/6$. If this is the case, this may also help to understand why our k_2/k_1 ratio is much smaller than the predicted value of 11.7 [44]. On the other hand, the weak p -variation of T^* explains the difference in the observed scaling behavior between here

and Ref. [29] where the CF effect is absent.

To our knowledge, there is currently no theory that can account for the observed features in $S(T)$, especially at low T . The most elaborated calculations by Zlatić *et al.* indicate that the sign change in $S(T)$ is a manifestation of the crossover from the weak-coupling local-moment regime to the strong-coupling Fermi-liquid regime with decreasing T [11]. While this is in a qualitative agreement with the experimental results at high T , it is difficult to reconcile the weak p -dependence of T^* and T_{\min} with the rapid growth of T_K over the broad p -range. Furthermore, the shape of $S(T)$ in the crossover region cannot be determined in their calculations so that a direct comparison with the scaling behavior is not possible. On the other hand, according to the semiphenomenological model developed by Fischer [31], the dominant contribution to negative $S(T)$ stems from interacting spin pairs. However, the model predicts that T^* should scale with T_K , which is at odds with our observations. Nevertheless, since the model considers only spin interactions, it will be interesting to investigate how the situation changes when the CF effect is included.

Another salient feature illustrated in Fig. 4 is that the large negative $S(T)$ minimum (the blue region) is located right above the superconducting domain up to almost p_c . A very similar situation is found when $S(T)/T$ is concerned [45]. Actually, as suggested in Fig. 3, this appears to be a common feature of the normal state of prototypical Ce-based p -induced superconductors. For CeAu₂Si₂, $|S_{\min}|$ and T_c exhibit a qualitatively similar p -dependence and hence one can speculate that this $S(T)$ minimum is intimately linked to SC. Here it is noted that T^* vanishes at a p considerably higher than p_c and shows a very different p -dependence from that of T_M , indicating that the negative $S(T)$ is not related straightforwardly to magnetic fluctuations. In this regard, the understanding of its physical origin may help to elucidate the pairing mechanism for these materials. Also, the scaling relation shown in Fig. 3 could serve as an empirical guide for the search of new Ce-based p -induced superconductors. For example, SC might be expected in Ce(Pt/Pd)Sn ($T_N \sim 7$ K) and CePb₃ ($T_N \sim 1.1$ K) under p , provided sufficiently high-quality crystals can be obtained.

Finally, it should be noted that substantial differences exist between the normal-state properties of CeAu₂Si₂ and CeCu₂X₂ despite their similarities [12]. In particular, regardless its smaller overall Δ_{CF} , T^* of CeAu₂Si₂ (~ 120 - 150 K) is almost twice that of CeCu₂X₂ (~ 40 - 80 K). Moreover, the low- ρ_0 of CeAu₂Si₂ in comparison with CeCu₂Ge₂ suggests a longer mean free path, which is more favorable for unconventional SC [21]. Given that the interaction leading to the negative $S(T)$ may also be involved in the superconducting pairing, these factors could be the key to understanding the exotic ground-state properties of CeAu₂Si₂ under p .

IV. CONCLUSION

In summary, we have studied systematically the high- p thermopower and resistivity of CeAu₂Si₂ up to 27.8 GPa. For the first time, a scaling behavior is found in the $S(T)$ data below 20.5 GPa as a function of a reduced temperature T/T^* . The comparison with $\rho(T)$ results shows that the $S(T)$ magnitude is determined by the Kondo coupling J , while the CF splitting Δ_{CF} controls the characteristic temperatures of the sign change (T^*) and the negative minimum (T_{\min}). Up to almost p_c , a large negative $S(T)$ minimum regularly precedes the superconducting transition, suggesting that the two phenomena are closely related. Furthermore, the scaling relation is shown to hold up to $2T^*$ for $S(T)$ at ambient p of related systems with diverse crystal structures, testifying to its wide applicability. Our work demonstrates that thermopower can be useful in probing the p -evolution of CF energy scale in Ce-based KLS, but should be used with caution in estimating the T_K of these materials. This calls for new theoretical understandings.

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